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A tale of two lines: Searching for the $5s-5p$ resonance lines in Pm-like ion spectra

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Abstract.
Highly charged ions in the promethium sequence have been suggested to show spectral features resembling the alkali sequence ions. Guided by calculations, the $5s-5p$ resonance lines have been sought in a variety of experiments. In the light of the most extensive calculations of Pm-like ions yet, applying relativistic multi-reference Møller-Plesset second-order perturbation theory, the experimental evidence is reviewed and the line identification problem assessed.

1. Introduction

In 1980, Curtis and Ellis [1] suggested on the basis of Hartree-Fock calculations that Pm-like ions (61 electrons) with a nuclear charge $Z > 73$ have alkali structure with a ground configuration $4f^{14}5s$. Dirac-Fock calculations by Theodosiou and Raftopoulos [2] added important relativistic corrections and then predicted the turning point at which $4f^{14}5s\ ^2\mathrm{S}_{1/2}$ rather than $4f^{13}5s^2\ ^2\mathrm{F}^o_{7/2}$ becomes the ground state to lie near $Z = 77$ instead of $Z = 74$. Since then, the prospective $5s-5p$ resonance transitions have been searched for in the spectra of tokamaks, laser-produced plasmas, vacuum sparks, foil-excited ion beams, ion-atom collisions, and electron beam ion traps. A number of identifications of these lines in Pm-like ions of W ($Z = 74$) through U ($Z = 92$) have been proposed, but at wavelengths that are neither in agreement with theory nor consistent within the data set. We have undertaken calculations of the term energies in Pm-like ions using second-order single-reference Møller-Plesset (MP2) and multi-reference Møller-Plesset (MR-MP) theory, both as a demonstration of the capabilities of our calculational approach and in order to compare with the experimental observations. Our computational procedures have been described elsewhere [3, 4, 5, 6].

One of our results is that the $4f^{14}5s\ ^2\mathrm{S}_{1/2}$ level is the ground state from Pt ($Z = 78$) onwards. In W, an element of high interest for the ITER fusion reactor experiment [7, 8], the $5s$ level is only the 24^{th} lowest level - far above the true ground state $4f^{13}5s^2\ ^2\mathrm{F}_{7/2}$. Similarly, the $4f^{14}5p$ states are actually not the lowest odd-parity levels in the ions up to Pb^{21+}, the levels being affected by numerous crossings with $4f^{12}5s^25p$ levels. The admixture of those states is so significant that it requires an extensive multi-reference treatment of the correlation effects in the theoretical approximations. This finding immediately suggests that the lines of primary interest might not be standing out as the resonance in the true alkali sequence spectra do, but be accompanied by many others of comparable, if not higher, intensity.

2. Review of experiments

When wide humps were observed in the EUV spectra of fusion plasma devices like ORMAK at Oak Ridge [9], a search started for signature lines of Pm-like ions that would in turn reveal the charge state distribution and thus the temperature of such plasmas. A beam-foil experiment (with fast Au ($Z = 79$) ions) at Brookhaven [10] remained inconclusive. A sequence of follow-up experiments at Bochum [11, 12] found signature lines in neighbouring lower charge states, and clusters of partly resolved lines that were believed to arise from the many core-excited states of Pm-like ions. Among the weak lines nearby are candidates for the $5s - 5p$ transitions, at wavelengths just short of various predictions.

Hutton *et al.* [13] have reported on identifications of two $5s-5p$ lines in the Pm-like spectrum of W ($Z = 74$) as observed in the Berlin electron beam ion trap (EBIT). Such EBITs are low-density devices, and the core excited level decays that swamp the beam-foil spectra are not expected to show at all. The charge state distribution in an EBIT can be varied by adjusting the electron beam energy. Based on such a variation, a number of lines were given charge state identifications or even line labels; however, without any known reference line in any of the tungsten spectra, such a result remains doubtful. The wavelengths of the lines identified with the $5s - 5p$ transitions differ from prediction by clearly different amounts, which signals consistency problems. Quite possibly no more than one of the two lines is from Pm-like W, if any.

Hutton *et al.* [13] have reported one line ($5s - 5p_{1/2}$) from a beam-foil experiment on Pb ($Z = 82$) done at RIKEN (Japan), with a wavelength a little shorter than predicted, just as observed with the candidate lines in the Bochum beam-foil spectrum. The spectrum is shown in a recent paper by Wu and Hutton [14].

Uranium in a (low density) tokamak plasma [15] is the heaviest element of which a line has been assigned to the $5s - 5p_{3/2}$ transition in a Pm-like ion so far, at a wavelength slightly longer than predicted. Unfortunately, in a tokamak there is no reliable way of actually determining the charge state of a highly charged ion, and hence the proximity of the selected line to the calculated wavelength of a specific ion transition may be fortuitous. The authors of ref. [15] note that, according to their collisional-radiative model calculations, the Pm-like ion resonance lines have to be accompanied by stronger lines from the $4d^{13}5s^2$ -$5s5p$ array and other, similar satellites that were not identified in the experiment.

3. Discussion

In short, although identifications of $5s - 5p$ lines of Pm-like ions have been suggested for W, Au, Pb, and U, the evidence is meagre. To our present judgment, none of the experimental investigations has unequivocally demonstrated seeing the $5s-5p$ "alkali-metal like ion resonance lines" yet. Evidently, the set of suggested identifications presented by Hutton *et al.* [13] is not intrinsically consistent and therefore hardly convincing; of the six lines used by Hutton *et al.* for a comparison to theory, five are known to be wrongly associated with $4f^{14}5s - 5p$ transitions or at least strongly suspected to be. Apparently there are only three line identifications that are not yet discounted; all three are from beam-foil spectra: two lines of Au [12] and one of Pb [13]. The three experimental wavelengths are about 1% shorter than our calculations (see [6]) predict. That is a poorer agreement than we have achieved in our work on fewer-electron ions. However, the three experimental wavelengths are not particularly accurate: the 1 Å measurement uncertainty corresponds to about 0.3 to 0.5%. There certainly is room for improvement on the experimental side.

In the present context, the best spectra (with isolated lines and the least clutter by unresolved core-excited state contributions) are those obtained at the Berlin EBIT [13]. However, our calculations confirm the much earlier calculations by Theodosiou and Raftopoulos [2] in that the spectrum of Pm-like ions of W has practically no alkali-like character, and the $5s - 5p$ transitions cannot stand out at this low ion charge. The question arises as to what actually has been seen at the Berlin EBIT. Simulated Pm-like W spectra, modeled once for high density (based on our MR-MP calculations) and once for low density (EBIT) conditions (based on a collisional-radiative model using the FAC code [16]) are shown in figure 1. The low-density model spectrum resembles the Berlin observations, but holds amost as many lines of ions of a single charge state as the EBIT spectra that surely represent a charge state distribution of several charge states of tungsten (in addition to lines from He). Calculations of the level structure and spectra of W^{7+} through W^{13+} [17] indicate a number of possible line identifications. It seems likely that among the lines seen are some of the Pm-like ion, but the $5s - 5p$ transitions are not prominent among them.

For a meaningful analysis of the W spectra obtained at the Berlin EBIT, a more complete set of observations would be helpful. Moreover, the calculations have to evolve towards matching the configurations and levels treated in accurate calculations (like our MR-MP) with those of collisional-radiative models based on FAC. However, many levels cannot be identified easily, because the total angular momentum of some of the electrons is no longer a good quantum number. The FAC code has proven to be robust and versatile. The accuracy of the FAC level energies, however, is not sufficient for spectroscopic applications within a given atomic shell. Considering that hundreds of levels have to be included in the model output, the substitution of accurate MR-MP level energies into FAC will have to be automated.

Concerning the experimental pursuit of alkali-ness in Pm-like spectra, the most viable option is provided by electron beam ion traps. This device offers an adjustable charge balance similar to the beam-foil interaction process, but EBITs work better for more highly charged ions. Studies in search for the $5s - 5p$ transitions outside a closed-shell core in alkali-metal like ions would best start with U and then continue towards lighter elements. Pm-like U^{31+} with its ionization potential of only about 1.1 keV is accessible at all present-day EBITs.

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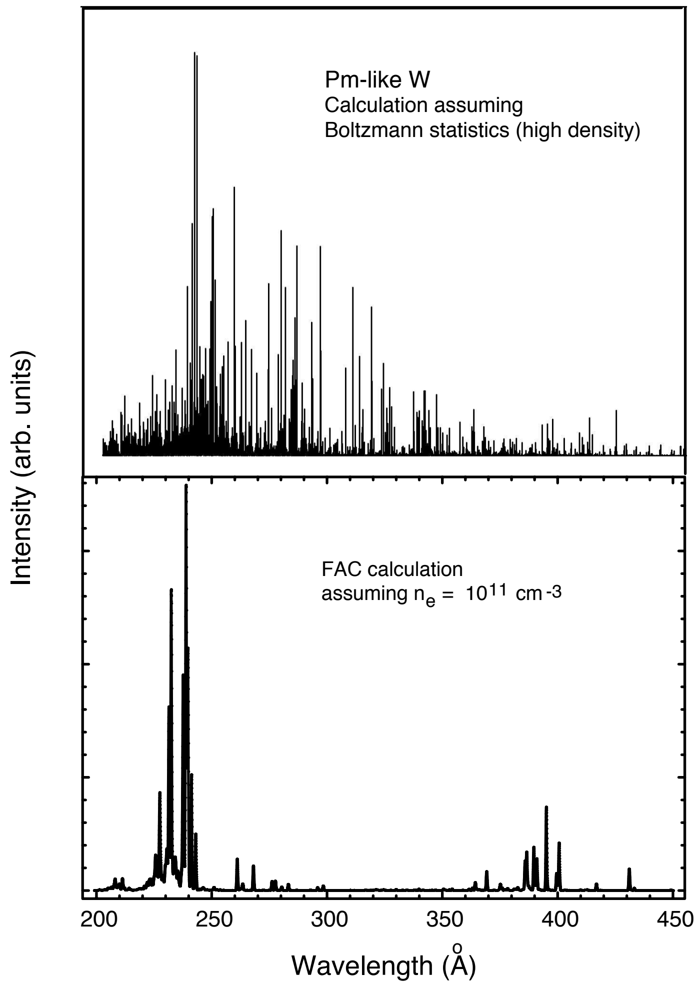

Figure 1. Simulated spectrum of the Pm-like ion W^{13+} in the range of the Berlin EBIT spectra shown by Hutton *et al.* [13, 14]. The upper simulation is based on the present MR-MP calculation and assumes high electron density (Boltzmann level population). The lower spectrum (courtesy of M F Gu (Livermore)) is based on FAC code calculations, for a low-density plasma ($n_e = 10^{11}\,\mathrm{cm}^{-3}$) as is typical for an EBIT.

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